‘Ordering’ in glasses and melts: Structural observations and their properties implications

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As the chemical and physical behavior of magmas dominates many geological processes, and as most technological glasses and glass ceramics start off in the molten state, understanding of the physical chemistry of silicate glasses and melts is important in both the earth and material sciences. Our present knowledge of these systems is limited, however, by the technical difficulties of working at high temperatures, and the theoretical complexities of materials whose structure and dynamics lie somewhere between the relatively well-understood field of organic polymers and that of molten salts. One of the most important questions concerns the link between macroscopic properties and structure at the nano-scales of the glassy state but also of the liquid and in the melts with changing intensive properties.

One of the more challenging and controversial idea is about middle and long range order in glass. Greaves *et al*. [1981] and Greaves [1985] proposed the *Modified Random Network* (MRN) model, where a glass can be a mixing of between rich domain in network former and rich domain with cation modifier that create some channels. This assumption can be used to explain alkali diffusion in glass [Roling *et al*. 2001; Sunyer *et al*. 2002; Meyer *et al*. 2004], and even if EXAFS experiments were over interpreted to build this original structural view, the channel idea works well to explain glass structure.

Following this idea, a glass is a non-random structure composed of two interpenetrated sub-networks: one rich in network former, surrounded by the other rich in network modifier or charge compensator, concentrated in channels already observed by Frischat *et al*. [2004] using high resolution AFM on Duran® borosilicate glass surface. These two different domains can be also interpret in term of low and high density region following Tanaka works to explain fracture, unmixing, polyamorphism and crystallization in glass [Kurita and Tanaka, 2004; Shintani and Tanaka, 2006; Patrick Royall *et al*. 2008; Furukawa and Tanaka, 2009].

We plan to discuss and complete this idea using some new rheological and structural investigation on alkali aluminosilicate glasses and melts. These considerations are of prime importance for glasses industry, earth sciences and more particular for explosive volcanos activity.

Volatile and major element zonation within melt inclusions: A natural diffusion experiment

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The diffusivities of volatile elements in silicate melts significantly impact petrological processes [e.g. 1, 2]. Although many studies of volatile diffusion in silicic melts have been undertaken, there have been few studies in basaltic melts [e.g. 3], and most of these have concentrated on the diffusion of only one or two elements in each experiment.

Significant zonation in volatile and major elements has been discovered in olivine-hosted melt inclusions from the Siqueiros Fracture Zone [samples previously studied in 4]. Elements that are compatible in olivine, such as Mg and Fe, are depleted at the edges of the zoned melt inclusions relative to their centers, whereas elements that are incompatible in olivine, such as Al, Ca, Si, Na and S, are enriched. Preservation of this zonation suggests that it formed during crystallization of olivine on the walls of the inclusions in response to cooling during or just prior to eruption. Although all melt inclusions from this sample suite exhibit zoning profiles, there are few previous reports [e.g. 5] of comparable concentration profiles in melt inclusions, so it is not known if this feature is widespread.

The MgO concentration profiles in two melt inclusions have been used to constrain the cooling history of the inclusions using the results of [6]. The profiles are consistent with an initial slow cooling rate followed by a period of more rapid cooling. For other components, their zoning profiles can be used to estimate their diffusivities relative to that of MgO. Of the volatile elements, we have demonstrated such profiles in S, and have preliminary results on Cl and F that allow constraints to be placed on their relative diffusivities in basaltic melt.

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